

REMARKS**Claims Amendment**

Claims 13 has been amended to more particularly point out and define the claimed invention. Support for the amendment to Claim 13 is found on page 6, lines 19-21 (“inter-penetrating network”), and on page 31, line 37 through page 32, line 5 of the English translation of the instant application (“between 95% and 10% by weight of polyvinyl-phosphonic acid” and “between 99.5% and 3% by weight of polyvinyl-phosphonic acid”).

Claim 14 has been amended to more particularly define and point out the present invention. Amendment to Claim 14 is supported by the disclosure on page 7, lines 26-27 of the English translation of the instant application (“at least 10%”).

Claim 21 has been amended to more particularly point out and define the claimed invention and to correspond in scope to Claim 13 as amended. Support for the amendment to Claim 21 is found on page 31, line 37 through page 32, line 5 of the English translation of the instant application.

New Claims 33 - 34 have been added to specific embodiments of the present invention. Support for Claims 33-34 is found on page 27, lines 6-13 (“at least 50% by weight”, or “at least 70% by weight” of the vinyl-containing phosphonic acid) of the English specification as filed.

Additionally, Claims 13 and 21 have been amended to recite the term “polyvinyl-containing phosphonic acid” instead of the term “polyvinyl-phosphonic acid”. These amendments are supported by an observation obvious to one of ordinary skill in the art that a vinyl-containing phosphonic acid polymerizes into a polyvinyl-containing phosphonic acid.

Applicants’ Correction of Previously Made Statements

In their response to an Office Action filed on November 29, 2007, Applicants stated, referring to U.S. 6,607,856 (“Suzuki”):

... nothing in Suzuki teaches or suggests the use of *in situ* polymerization of monomer within a polymer matrix.

In making this statement, Applicants inadvertently failed to appreciate the teachings of Example 18 of Suzuki (column 29, lines 44-64). Applicants regret any confusion this may have caused. This error was made without deceptive intent.

Applicants' Invention

Applicants' invention is a proton-conducting electrolyte membrane obtained by a method recited in Claim 13, as amended, and new Claims 33-34. The steps include (a) expanding a polymer film with a liquid that contains a vinyl-containing phosphonic acid; and (b) polymerizing the vinyl-containing phosphonic acid present in the liquid of step (a), thereby forming an interpenetrating network of polyvinyl-phosphonic acid and the polymer.

Claims 13, and 33-34 further specify that the product obtained in step (b) includes at least 10%, between 10% and 95%, at least 50%, or at least 70% by weight of vinyl-containing phosphonic acid, respectively. The recited lower limits on the percent weight of vinyl-containing phosphonic acid ensure that the intrinsic conductivity of the inventive membrane at 160°C is at least 0.001 S/cm. (See the English translation of the specification as filed: Table 1, page 41; Table 2, page 42; Table 3, page 43; Table 4, page 44; Table 5, page 45; Table 6, page 46; and Table 7a, page 47.)

In other words, the process steps recited in the base claims result in well-defined material features (percent weight of vinyl-containing phosphonic acid, an interpenetrating network of polyvinyl-phosphonic acid and the polymer). These material features, in turn, confer unexpected advantages on the proton-conducting electrolyte membrane obtained by the method recited in the base claims that could not have been predicted based on the cited references.

Rejection of Claims 13-16 and 18-22 under 35 U.S.C. §§102(b), 102(a), 102(e) or 103(a) over Zouahri et al.

Applicants previously argued that the date of acceptance of Zouahri's publication, March 20, 2002, is after the earliest priority date of the present application, March 5, 2002. As such, Zouahri *et al.* is unavailable as a prior art reference.

In the Office Action mailed on February 14, 2008, the Examiner stated that a claim of priority under 35 U.S.C. §119 that Applicants assert must be perfected by filing a certified English translation of the priority document with the USPTO.

Applicants submit herewith a certified English translation of the German Patent No. 102 09 419.5, which is the priority document of PCT/EP03/02399, of which the instant U.S. utility application is a U.S. national stage. As can be seen, Claim 1 of German Patent No. 102 09 419.5, which corresponds to Claim 13 of the instant application, is supported by the disclosure of the priority document. As such, base Claim 13 is entitled to the priority, rendering Zouahri's publication unavailable as prior art.

Reconsideration and withdrawal of the rejection are respectfully requested.

Rejection of Claims 13-16 and 18-22 under 35 U.S.C. §§102(b), 102(a), 102(e) or 103(a) over U.S. 5,643,968 ("Andreola")

Applicants previously argued that there is only one polymer in the composition of Andreola, namely, the one in which an ion exchange functionality (*i.e.* a charged group) is grafted onto an aromatic backbone. Applicants further argued that the claimed electrolyte membranes comprise an inter-penetrating network of two independent polymers.

In the Office Action mailed on February 14, 2008, the Examiner stated that this argument was not compelling as the feature of "inter-penetrating network" was not recited in the claims.

Applicants amended base Claim 13 to recite that "polymerizing the vinyl-containing phosphonic acid present in the liquid of step a) [...forms] an interpenetrating network of polyvinyl-phosphonic acid and the polymer". Similar recitations appear in new Claims 33-34. Thus, as Applicants previously argued, the membranes that results from performing the steps recited in Claim 13 are materially different from the material disclosed by Andreola. This material difference is now reflected in the claim language.

Reconsideration and withdrawal of the rejection are respectfully requested.

Rejection of Claims 13-16 and 18-22 under 35 U.S.C. §§102(b), 102(a), 102(e) or 103(a) over U.S. 6,248,469 ("Formato")

Applicants previously argued that the substrate polymer of Formato is materially different from the polymer film of step (a) of Applicants' Claim 13, and that, therefore, the composite membranes resulting from the methods of manufacturing described by Formato are also materially different from the membrane of Claim 13.

With reference to FIG. 1 of Formato, Applicants argued that the porous regions of the final product of Formato (*i.e.* the regions in which "voids" were formed by solvent leaching) are separate and distinct from polymer regions (*i.e.* regions in which *no* "voids" were formed). Applicants further argued that, in the final product of Formato, the ion-conducting material is in the porous regions, and not in the polymer regions. (Applicants particularly noted that the two types of regions are expressly marked by different labels in panel C of FIG. 1.) Applicants finally argued that the membrane of Formato, even if prepared by employing *in situ* polymerization of the ion-conducting material, will have bulk regions in which little or no ion-conducting material is present. This is materially different from the membranes of the present invention, which form an inter-penetrating network of different polymer *molecules* (rather than a network of different *regions*, as in Formato).

In the Office Action mailed on February 14, the Examiner stated that (1) the pending claims do not specifically preclude the presence of voids; (2) Formato expressly teaches that an inter-penetrating network is formed (Formato, col. 17, ll. 22-44); and (3) the process by which the resulting product is made is not relevant to the issue of patentability of said product.

In response, Applicants first note that the differences between the processes of making the product of Formato and the claimed membranes have not been relied upon by Applicants. Applicants again direct the Examiner's attention that the *product* of Formato is described as being *materially different* from the membranes formed by the process recited in Claim 13 due to presence of bulk regions in which little or no ion-conducting polymer is present.

Regarding Formato's teachings that an inter-penetrating network was formed within his product, Applicants note that the term "inter-penetrating network" refers to a *microscopic* property of mixtures of polymers: covalent or non-covalent crosslinking of polymer backbones. (To further support this statement, Applicants present herewith Exhibit A: a printout of a web

page “Organic-Inorganic Interpenetrating Networks”, available at URL <http://www.psrc.usm.edu/mauritz/nano4.html>, last accessed on July 30, 2008, maintained by School of Polymers and High Performance Materials of University of Southern Mississippi, Department of Polymer Science, the Research Group of Dr. Mauritz.) Formato, in contrast, teaches a network of *macroscopic* “channels” containing ion-conducting polymer and penetrating a bulk of non-conducting matrix. Formato’s material does not comprise an “interpenetrating network” within the meaning of this term as understood by one of ordinary skill.

Regarding the Examiner’s assertion that the pending claims do not exclude porous materials, Applicants direct the Examiner to M.P.E.P. §2113, which states regarding product-by-process claims:

The structure implied by the process steps should be considered when assessing the patentability of product-by-process claims over the prior art [...] (*Emphasis added.*)

The structure implied in Claim 13, *i.e.*, the structure inevitably formed by the process recited in Claim 13, which is a tightly woven network of two polymer backbones, does not include pores of the type present in the compositions disclosed in Formato. Formato teaches filling a “sponge”. No entangling of polymer backbones (*i.e.* creation if an interpenetrating network) occurs by filling channels, pores and cavities in a substrate. Therefore, the claimed product is novel and non-obvious in view of Formato, even though the absence of these pores is not recited in the claims.

Membranes produced by the processes recited in Claim 13, as amended, and new Claims 33-34 are patentable over Formato’s products for at least one more reason, *i.e.*, the presence in the membranes of the present invention of at least the specified percentage by weight of vinyl-containing phosphonic acid. As noted above, the presence of at least 10% by weight of vinyl-containing phosphonic acid confers the unexpected advantage, *i.e.*, high conductivity at elevated temperature.

In summary, Formato does not disclose a material having the features listed above. As such, Formato does not anticipate the inventions of Claims 13, as amended, and new Claims 33-34. Moreover, Claim 13, as amended, and Claims 33-34 are also non-obvious over Formato because the features implied or recited by the process of the base claims confer onto the

membranes of the present invention an unexpected advantage of high intrinsic conductivity of the inventive membrane at elevated temperatures.

Reconsideration and withdrawal of the rejection are respectfully requested.

Claim 13, as Amended, and new Claims 33 - 34 are Novel and Non-Obvious in view of U.S. 6,607,856 ("Suzuki")

Suzuki fails to either anticipate the pending base claims or to render these claims obvious.

First, nowhere does Suzuki provide an example of an *in situ* polymerization in which the "doping ratio" (*i.e.* the percent by weight of the acid-carrying polymers added to a primary polymer material) was at least 10% by weight. As such, Claim 13, as amended, as well as new Claims 33-34 are novel over Suzuki.

Secondly, as Applicants argued above, the recited lower limits on the percent weight of vinyl-containing phosphonic acid ensure that the intrinsic conductivity of the inventive membrane at temperatures of 160°C is at least 0.001 S/cm.

Thus, Applicants' membranes possess unexpected advantages not disclosed or suggested by Suzuki. Specifically, Applicants' membranes show conductivity at very high temperatures, above the boiling point of water, and thus can function without moistening. (See page 5, lines 29-31, and the paragraph bridging pages 5 and 6 of the English translation of the instant Application). In fact, the Applicants' membranes retain conductivity at 160 °C, at which temperature no water is present (see Tables in Examples 1-27, pages 40-50, which show that various embodiments of the membranes of the present invention possesses high conductivity at 160 °C). This is to be contrasted with the materials of Suzuki, which only show conductivity in the presence of water (see Suzuki, column 14, lines 29-40).

Reconsideration and withdrawal of the rejection are respectfully requested.

CONCLUSION

In view of the above remarks, it is believed that all claims are in condition for allowance, and it is respectfully requested that the application be passed to issue. If the Examiner feels that a telephone conference would expedite prosecution of this case, the Examiner is invited to call the undersigned.

Respectfully submitted,

HAMILTON, BROOK, SMITH & REYNOLDS,
P.C.

By Alexander Akhiezer

Alexander Akhiezer

Registration No. 54,617

Telephone: (978) 341-0036

Facsimile: (978) 341-0136

Concord, MA 01742-9133

Dated:

8/12/03

Organic-Inorganic Interpenetrating Networks

An interpenetrating polymer network (IPN) is any material containing two polymers, each in network form. The three conditions for eligibility as an IPN are: (1) the two polymers are synthesized and/or crosslinked in the presence of the other, (2) the two polymers have similar kinetics, and (3) the two polymers are not dramatically phase separated. Of course, these are loosely held guidelines. IPNs that have only one polymer crosslinked (where the polymers are synthesized separately) or where the polymers have vastly different kinetics are still considered to be IPNs. IPNs are distinguishable from blends, block copolymers, and graft copolymers in two ways: (1) an IPN swells but does not dissolve in solvents, and (2) creep and flow are suppressed.

Several kinds of IPN architectures exist (See Figure 1). These systems differ mainly because of the number and types of crosslinks that exist in the system. A non-covalent semi-IPN is one in which only one of the polymer systems is crosslinked. A

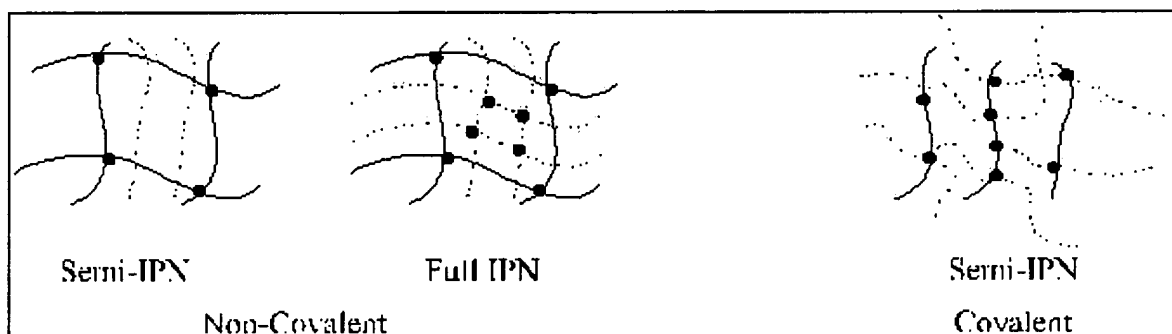


Figure 1: IPN Materials

non-covalent full IPN is one in which the two separate polymers are independently crosslinked. A covalent semi-IPN contains two separate polymer systems that are crosslinked to form a single polymer network. This covalent semi-IPN is similar to a non-covalent IPN because one of the polymer systems can be crosslinked without networking with the second linear system. However, the two systems tend to be networked for better property development. These covalent semi-IPNs are developed with organic-inorganic composite materials.

Non-Covalent IPN Materials

A wide variety of organic-inorganic non-covalent IPN materials have been formulated in an effort to improve material properties. Inorganic incorporation into polymers ranging from polyacrylates and polyesters to polyimides and nylons have been attempted. The problem with the non-covalent systems, which can also be a problem with the covalent systems, is the lack of an effective interface. This problem could stem from several factors including surface energy phenomena and lack of molecular interactions between phases. Figure 1 in Organic-Inorganic Associations shows several polymers that can interact with the inorganic phase. These polymers are proposed to hydrogen bond with the inorganic phase, creating an interface between the two materials (See Figure 2). However, the key to having non-covalent

organic-inorganic materials is not only utilizing a polymer that can have hydrogen bonding between the two phases but also to have low loading of the inorganic phase. Low loading of the inorganic phase will result in an increase in the overall material properties without sacrificing the interfacial bonding.

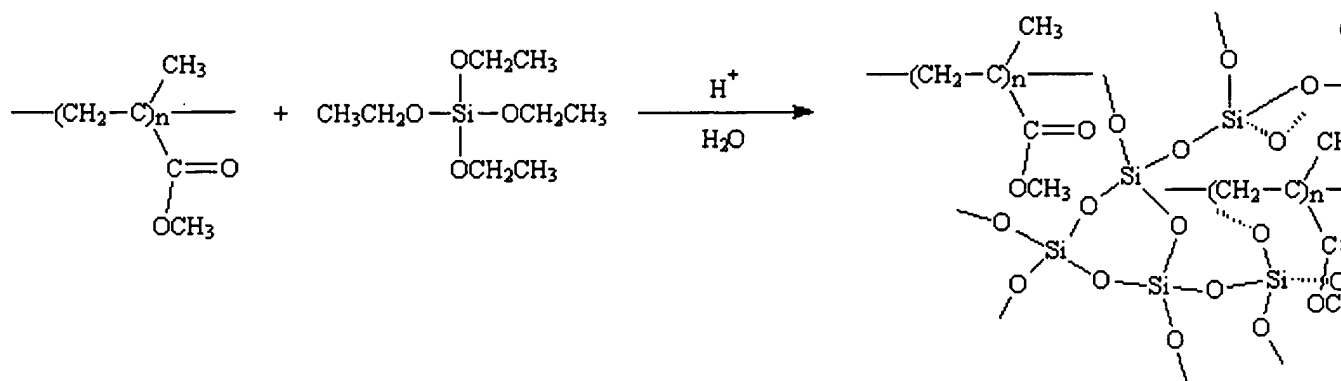


Figure 2: Non-Covalent IPN Materials

Covalent IPN Materials

A variety of polymers and copolymers have been synthesized incorporating reactive silicon alkoxides along the backbone of the polymer (See Figure 3). Silicon alkoxide incorporation into the polymer backbone can be accomplished with many monomers and through various synthetic means (See Figure 4). For example, silicon alkoxides can be incorporated into a polymer backbone via free-radical polymerization through a vinyl moiety, via a condensation reaction with an organic moiety on the silicon alkoxide monomer, or via a post-reaction (such as a hydrosilylation reaction). Through covalent attachment of the reactive silicon alkoxides, polymer-polymer interfacial

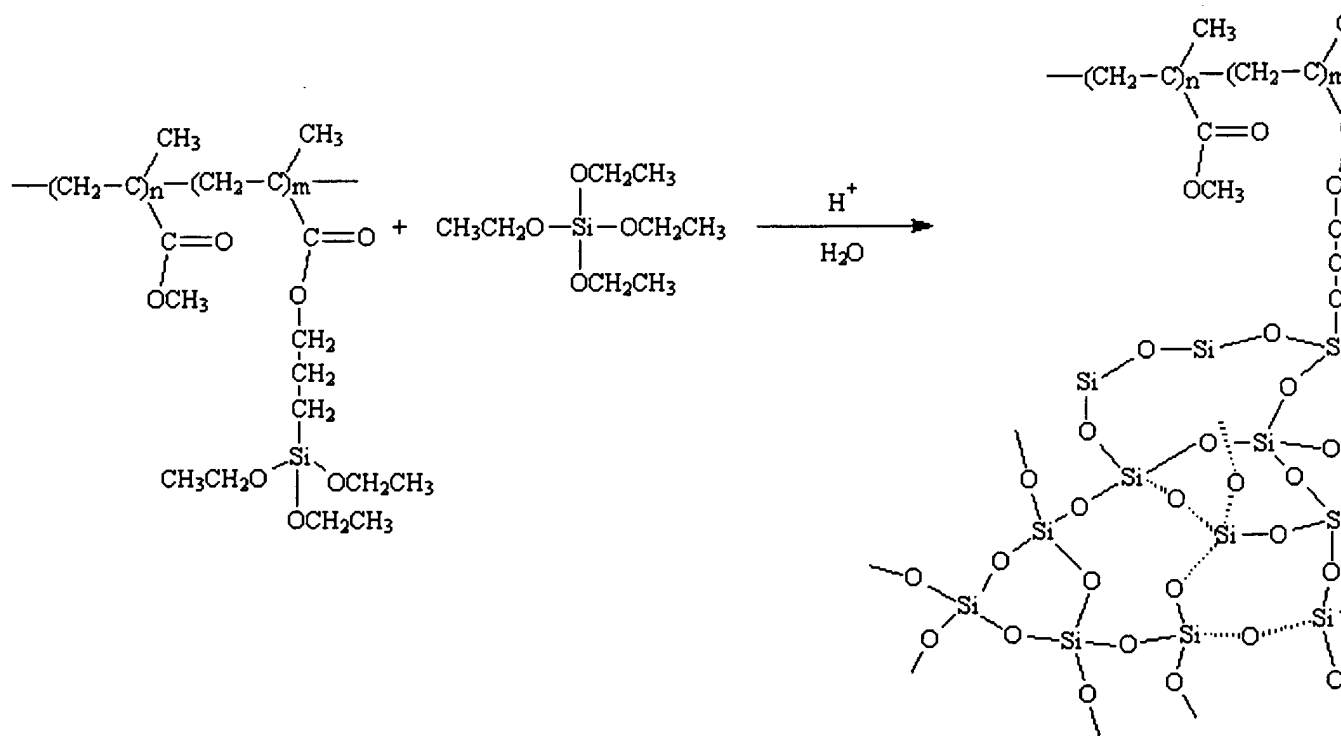
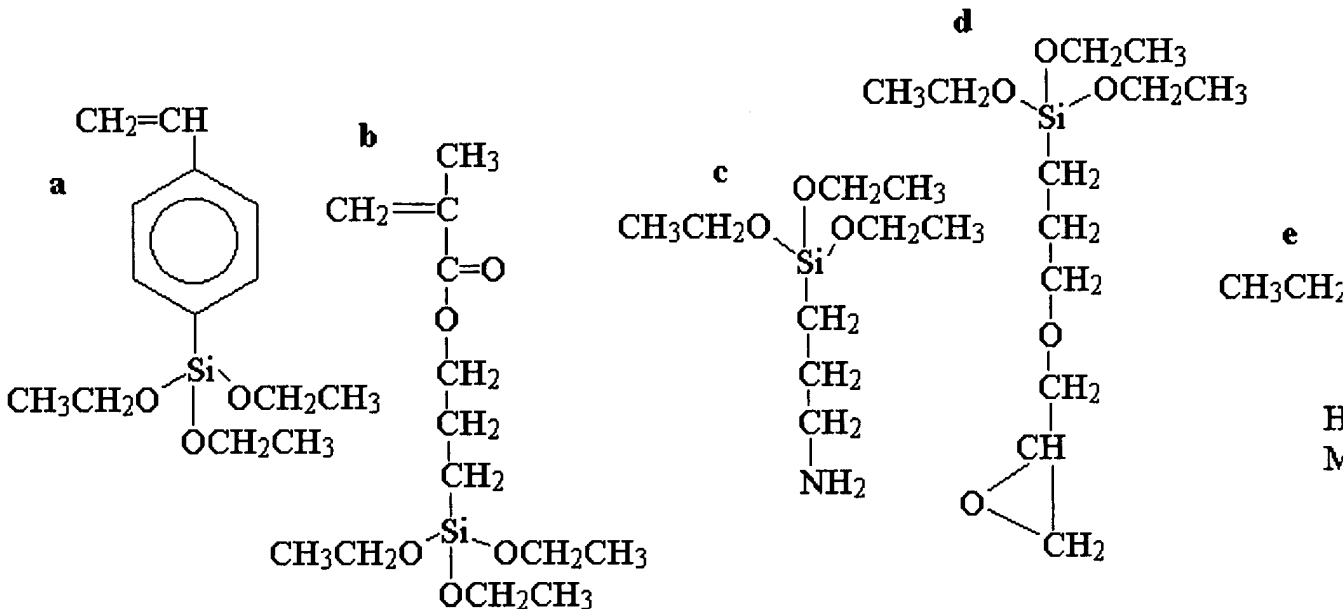


Figure 3: Covalent IPN Materials

problems were hypothesized to be reduced. However, covalent IPN materials can have similar problems with the interface as the non-covalent materials. Again, similar to the non-covalent systems, a general lack of cohesiveness between the two phases can exist at molecular weight loadings higher than 10%. This problem with the gross phase



Vinyl Polymerization Monomers (a,b)

Condensation Monomers (c,d)



Figure 4: Some Monomers Utilized in Covalent IPN Materials

separation at the interface is under investigation by researchers. Utilization of a variety of intermolecular bonding forces seems to improve upon the overall separation problems of the material. For example, by utilizing a polymer with a covalently bound silicate material that can also hydrogen bond with the organic polymer backbone creates more opportunities for better interfacial interactions.

References:

7. Sperling, L. H. "Interpenetrating Polymer Networks and Related Materials", Plenum Press, 1981, Chpt. 1.

Written by: Sandra Young (Partially From Her Research Prospectus)

-  **Go back to the site index page**
-  **Go back to the nanocomposites page**



Questions or comments about this page can be mailed to Dr. Ken Mauritz at Kenneth.Mauritz@usm.edu